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Measurement and control of attosecond pulses

Paul Corkum
NATIONAL RESEARCH COUNCIL OF CANADA

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Aim 1. To measure simultaneously the spatial and the temporal structure of an isolated attosecond pulse while also simplifying attosecond metrology so it becomes more widely accessible. Result: This goal was completely accomplished.
Aim 2: To exploit few-cycle infrared radiation to create shorter duration attosecond pulses. Result: We currently reach photon energies > 500 eV but without phase control for the 1.8 μm driver.
Aim 3: To compare attosecond gating methods, selecting the most efficient way to produce the highest power or the shortest duration attosecond pulses. Results: We have adapted the attosecond lighthouse to gases and confirmed isolated attosecond pulse production with an IR driver.
Aim 4: To use the quantum path interference, present in any attosecond pulse, to resolve the strongly-driven attosecond time-scale electron dynamics in selected ions. Results: After discussions with the Program Manager, this aim have been modified to concentrate on harmonics generation in solids.

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Report on AFOSR project # FA9550-13-1-0010: *Measurement and Control of Attosecond Pulses*

PI: Paul Corkum
Joint Laboratory for Attosecond Science
University of Ottawa and National Research Council of Canada

This is a final report on the AFOSR project "Measurement and Control of Attosecond Pulses". The project began in January 2013 and terminated in December 2015.

Project goals.

1. *To measure simultaneously the spatial and the temporal structure of an isolated attosecond pulse while also simplifying attosecond metrology so it becomes more widely accessible.*

Result: This goal was completely accomplished.

Publications:

a. K.T. Kim, C. Zhang, A.D. Shiner, S.E. Kirkwood, E. Frumker, G. Gariépy, A. Naumov, D.M. Villeneuve and P.B. Corkum, "Manipulation of quantum paths for space-time characterization of attosecond pulses", *Nature Physics* **9**, 159-163 (2013)
b. K.T. Kim, C. Zhang, A.D. Shiner, B.E. Schmidt, F. Legare, D.M. Villeneuve and P. B. Corkum, "Petahertz optical oscilloscope", *Nature Photonics*, 7, 958-962 (2013)
c. K.T. Kim, D. M. Villeneuve, P. B. Corkum, "Manipulating quantum paths for novel attosecond measurement methods" *Nature Photonics* **8**, 187-194 (2014)
d. Yijian Meng, Chunmei Zhang, Claude Marceau, A. Yu. Naumov, P. B. Corkum, and D. M. Villeneuve, "Octave-spanning hyperspectral coherent diffractive imaging in the extreme ultraviolet range", *Optics Express* **23**, 28960 (2015)

Summary of key findings: Metrology has been a major challenge for attosecond science. Methods developed for optics cannot be directly extended, thereby making it necessary to develop new methods. The current "gold standard" for attosecond metrology, attosecond streaking, cannot be easily extended to low power attosecond pulses, or to very short wavelength pulses. Here, we introduced a flexible and comprehensive new metrology. It allows us to measure attosecond pulses with frequency extending to photon energies of 500 eV and beyond. This new metrology is highly sensitive, allowing weak pulses to be characterized for the first time while also providing the first space-time measurement ever made of an attosecond pulse. We find that attosecond pulses are always space-time coupled.

We call this form of measurement "in-situ" because the pulse is measured in the nonlinear medium as the pulse is being formed. While this may sound limiting, *any further modification of the spectral phase of any of the frequencies making up the pulse can be determined by spectral interferometry – a linear measurement*. Thus, when combined with spectral interferometry, in-situ measurement becomes a comprehensive measurement method.

The development of the method for isolated attosecond pulses is reported in reference [1 (a)] and reviewed in reference [1(c)]. Not only is the pulse measured, but simultaneously, we measure the weak perturbing field as reported in reference [1(b)].

The concept behind in-situ measurement is illustrated in Figure 1. A weak beam is incident on the nonlinear medium at a slight angle to the fundamental. It imposes a phased modification on the re-collision electron wave packet and therefore on the attosecond pulse that it produces. This modification deflects the beam and the deflection depends on the phase of the perturbing beam relative to the fundamental. As the relative phase changes, the attosecond beam changes direction. This change encodes the space-time structure of the pulse.

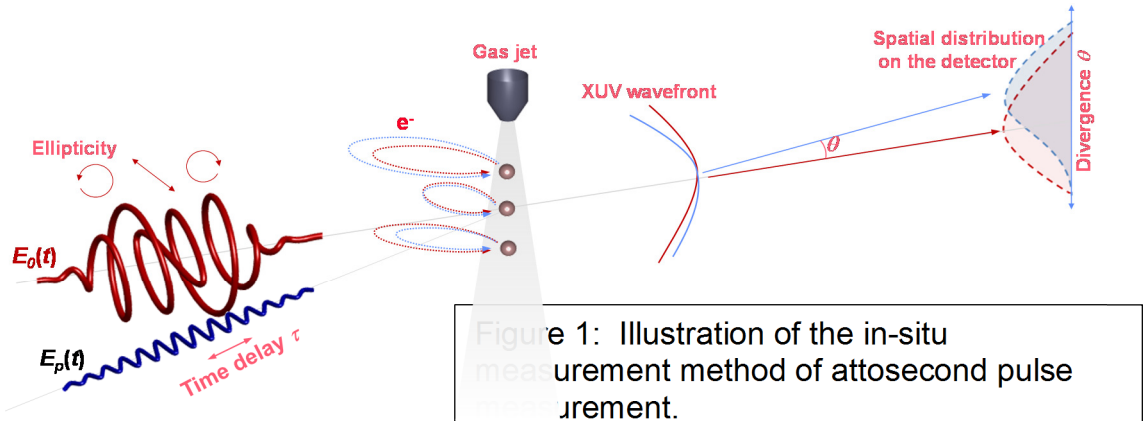
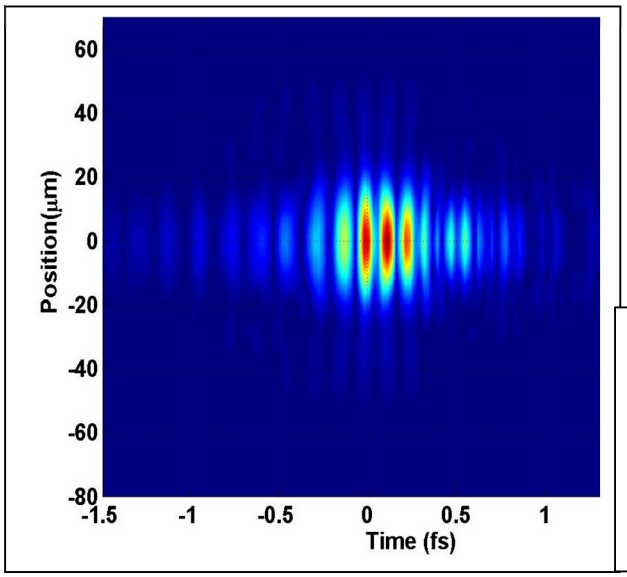


Figure 1: Illustration of the in-situ measurement method of attosecond pulse measurement.



We have applied the in-situ measurement method to the first measurement of an isolated attosecond pulse from an infrared (1.8 μm) driver. The space-time structure of the pulse is shown in Fig 2. The gating method was ultrafast wavefront rotation and the gas was argon.

Figure 2. The space (vertical axis) time (horizontal axis) of an isolated attosecond pulse produced by ultrafast wavefront rotation with a few cycle 1.8 μm pulse interacting with a jet of argon gas. Accepted for publication in Scientific Reports (2016)

As I write this report, we are generalizing the in-situ method to allow single-shot time-resolved pulse measurement.

Aim 2: *To exploit few-cycle infrared radiation to create shorter duration attosecond pulses.*

Result: We currently reach photon energies > 500 eV but without phase control for the 1.8 µm driver. We obtain CEP control with lower energy driver pulses, but with them we cannot reach 500 eV. Once we combine these, we expect to produce and characterize pulse with durations < 50 as.)

Publications:

a. A.D. Shiner, C. Trallero-Herrero, N. Kajumba, B.E. Schmidt, J.B. Bertrand, Kyung Taec Kim, H.-C. Bandulet, D. Comtois, J.-C Kieffer, D.M. Rayner, P.B Corkum, F. Légaré, D.M. Villeneuve, "High harmonic cut-off energy scaling and laser intensity measurement with a 1.8 µm laser source" Journal of Modern Optics, DOI:10.1080/09500340.2013.765067 (2013).
b. C. Zhang, G. Vampa, D.M. Villeneuve and P.B. Corkum, "Attosecond lighthouse driven by sub-two-cycle, 1.8 µm laser pulses" J. Phys. B.: At. Mol. Opt. Phys. 48, 061001 (2015).

Summary of key findings: Every method of generating isolated attosecond pulses requires carrier-envelop-phase control. We generate CEP controlled pulses for 1.8 µm light by using an OPA in which the seed, at the signal wavelength, is generated by continuum generation. In this way the idler is CEP stable. This pulse is then frequency broadened in a hollow core fiber and passed through fused quartz where anomalous dispersion compresses it. We achieve 600 µJ pulses with duration of less than 13 fs. With this source we can produce isolated attosecond pulses.

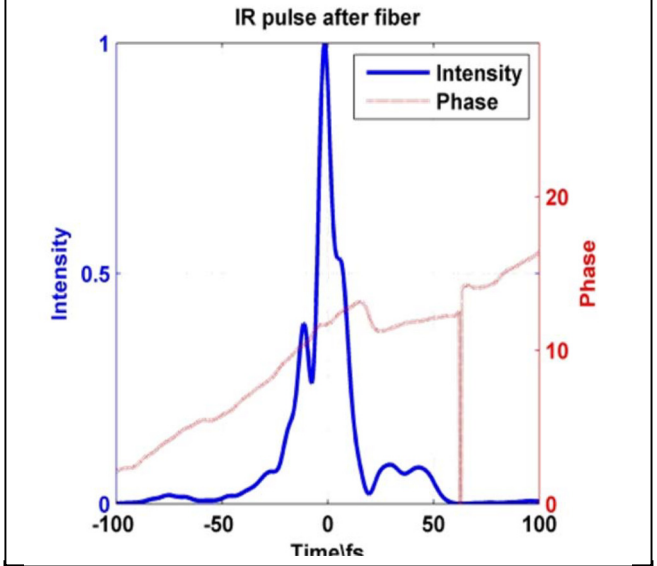


Figure 3: A FROG (Frequency Resolved Optical Gating) trace of a 1.8 µm beam as it leaves the hollow core optical fiber where it is spectrally broadened and the quartz plate where it is compressed.

With 600 µJ pulses we can reach the water window using neon gas and a thin jet with high backing pressure. In Ottawa we are unable to go further until we upgrade our laser system – a process that is just beginning. However in a related lab in Montreal we can make high energy pulses extending to greater than 500 eV photon energy.

As yet the Montreal system is not CEP stable. Once it is, we expect to reach pulses as short as 50 attoseconds, even without dispersion compensation.

Aim 3: *To compare attosecond gating methods, selecting the most efficient way to produce the highest power or the shortest duration attosecond pulses.*

Results: We have adapted the attosecond lighthouse to gases and confirmed isolated attosecond pulse production with an IR driver (to be published in Scientific Reports). We are working on new method using a solid irradiated at high intensity to create a space-time coupled pulses that then irradiate a gas jet.

Publications:

a. T.J. Hammond, K.T. Kim, C. Zhang, D.M. Villeneuve, and P.B. Corkum, "Controlling attosecond angular streaking with second harmonic radiation" *Optics Letters*, **40**, 1768, (2015).

Summary of key findings: We have explored two new gating methods. Both are promising. In one we use a fundamental beam that is space-time coupled. This creates a high harmonic train in the medium but each pulse in the train is travelling in a different direction. Therefore, as the beam propagates, it separates into independent pulses. The mechanism is illustrated in Fig 4.

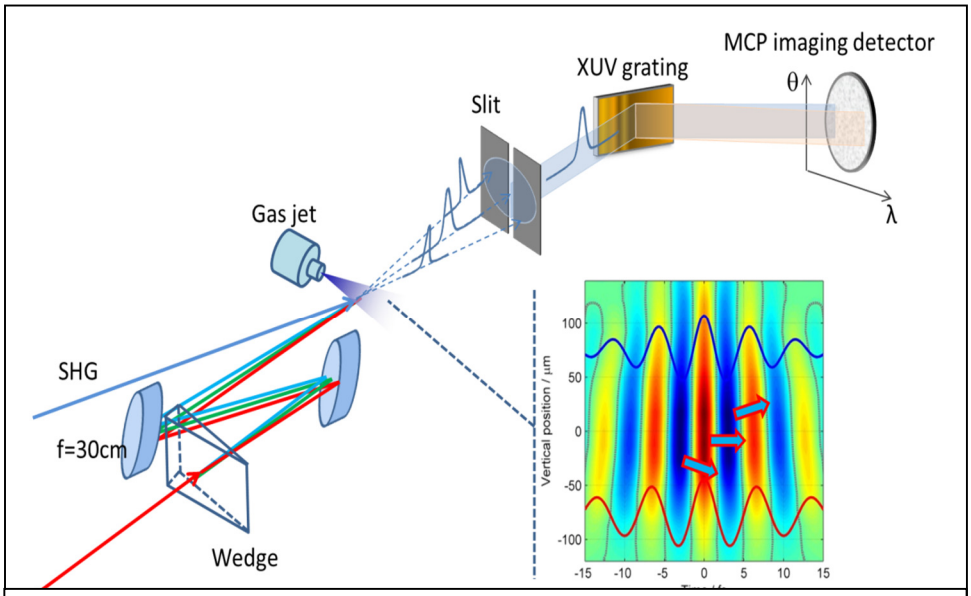


Figure 4. A fundamental beam passes through a thin wedge thereby slightly changing the direction of each color of light in the pulse. In the focus, where the gas jet is placed, the beam wave front rotates as shown in the inset. This creates a series of attosecond pulses. One of these pulses – an isolated attosecond pulse, can be selected by a slit in the far field and measured by streaking or by in-situ measurement.

A CEP stable few cycle pulse irradiates a thin wedge. Shown here is the arrangement that we used for a 1.8 μm fundamental beam.

This creates a beam at the focus that has a wave front that changes from one ½ cycle to the next. When

the fundamental beam is converted to harmonics the higher frequency of the harmonics means that the beam has lower divergence. The attosecond pulses that make up the harmonics, therefore separate into individual attosecond pulses in the far field.

The train of pulses created in this way are shown in the plot of angle of the radiation (vertical axis) as a function of the frequency. Figure 5(a) shows the three pulses created with the lighthouse method. Figure 5(b) shows a single pulse selected by an aperture in the far field.

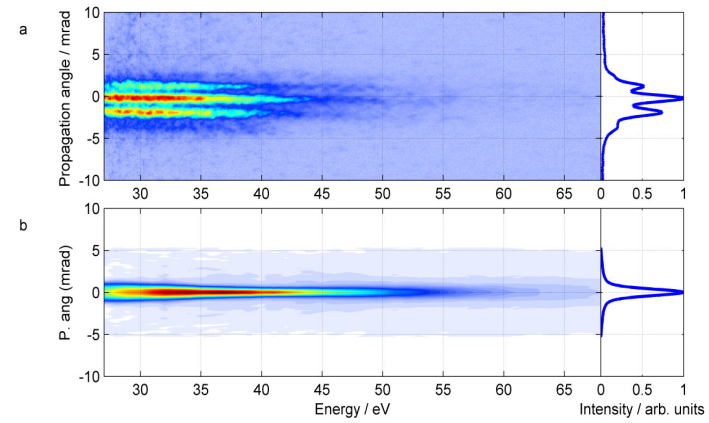


Figure 5: Beamlets as measured in the far field plotted with their angle of propagation shown for each frequency. Fig. 5 (a) shows the beamlets spread out along the direction of the wavefront rotation while Fig 5 (b) shows one beamlet that has been selected by the spectrometer slit when the wavefront rotation was perpendicular.

Figure 2 (page 2) is a space time resolved pulse obtained with argon gas. Krypton gas. It corresponds to the measurement in Fig. 5(b) except that Krypton gas was used for Fig 5.

We have extended this work neon and to the water window, but we have not measured the pulse duration.

The second gating method also uses a space-time coupled pulse. To create it we use of the response of solids to intense light. Since we do not yet have well prepared figures, I describe the method.

The fundamental pulse passes through a thin (about 200 μm) crystalline quartz plate. Its intensity should be greater than 10^{13} W/cm^2 but below about $4x10^{13}$ W/cm^2 where multi-pulse damage begins to appear. With a B integral of about 2 or 3, the beam begins to self-focus as it passes through the material and simultaneously creates second harmonic light. Since only one coherence length of the material can contribute to the second harmonic, the second harmonic signal is space time focussed in exactly the same way as the fundamental. With appropriate thickness of quartz, its phase can be tuned for double optical gating.

This space-time coupled, two-color pulse is then incident on a gas target placed some distance away where it creates an isolated attosecond pulse in Xenon, Krypton and probably all rare gases.

Aim 4: *To use the quantum path interference, present in any attosecond pulse, to resolve the strongly-driven attosecond time-scale electron dynamics in selected ions.*

Results: We have modified this aim to concentrate on solids. Therefore, we did not measure quantum path interference in the X-A transition in the N_2^+ ion. However, we are currently using strong field methods to study the X-A transition in N_2^+ using gain dynamics related to air lasing on the B to X transition.

Publications (related to the original aim):

a. J.B Bertrand, H.J. Wörner, P. Salières, D.M. Villeneuve and P.B. Corkum, "Linked attosecond phase interferometry for molecular frame measurements" *Nature Physics* 9, 174-178 (2013)

Summary of key findings: The issues that we proposed to study was to follow the dynamics between the X and A states of Nitrogen molecular ions as the molecule responds to the strong field during the first cycle following ionization. We expected to see sub-cycle dynamics in the field needed to ionize N_2. The observable was to be the interference pattern between the short and long trajectory electrons which originate at different re-collision times which we expected to change as we changed molecular alignment. We were hopeful for the experiment because in-situ measurement offered us very good resolution of the interference between these two trajectories. However the experiment required that we align the molecules. Combining alignment and in-situ measurement proved to be difficult.

The X-A transitions in N_2^+ is also important for creating inversion for air lasing. We now know that it is better to measure the X-A transition dynamics indirectly using gain dynamics on the B to X transition because X-A coupling will strongly influence the rotational wave packets that are imprinted on the gain. With that in mind, we are now studying the air lasing problem with the technology of high harmonic generation.

Furthermore, we began to understand the deep connection between attosecond pulses made in the gas phase and high harmonics formed in wide bandgap semiconducting material. After discussions with the contract monitor, we replaced Aim 4 with the aim to study solid state harmonics.

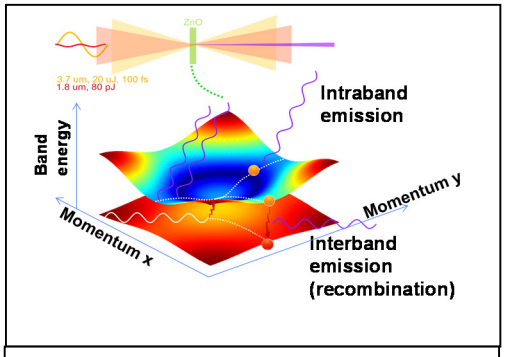


Figure 6: Illustration of a two band model describing the semiconducting properties of ZnO (from Vampa et al. Phys Rev Lett. **113** , 073901 (2014))

Publications related to high harmonics from solids:

b. G. Vampa, T.J. Hammond, N. Thiré, B.E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec and P.B. Corkum, "Linking high harmonics from gases and solids" *Nature* **522**, 462 (2015)

c. C.R. McDonald, G. Vampa, P.B. Corkum, and T. Brabec, "Interband Block oscillation , *Phys. Rev. A* **92**, 033845 (2015)

d. G. Vampa, T. J. Hammond, N. Thiré, B. E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec, D. D. Klug, and P. B. Corkum, "All-Optical Reconstruction of Crystal Band Structure", *Phys. Rev. Lett.* **115**, 193603 (2015)

By studying a two-band model of a wide bandgap semiconductors, we have discovered that, harmonics can have essentially the same origin in solids as in gases. The qualification "essentially" only refers to the natural generalization that electron and hole motion in solids takes the place of electrons and ions in gases and that the electrons and holes move with band corrected masses in a semiconductor rather than with their real electron and ion mass in gases.

Figure 7 (a and b) illustrates the dominance of re-collision harmonics (blue) in ZnO over the single band source (red). The single band source includes Block oscillations.

To experimentally test the theoretical prediction, we have performed a two color study of the high harmonics emission [4b]. (A two color study is closely related to the "in-situ" measurement methods described above.) The two color method was developed in my group and reported in 2006 in a paper in Nature Physics, **2**, 781 (2006) for gas harmonics. The spectral phase for a given emission frequency is encoded in the phase shift of the modulation of the closest even harmonics. The spectral phase is a measurement of the pulse duration of the associated attosecond pulse.

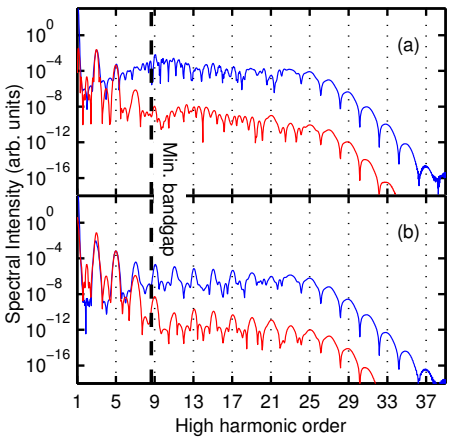


Figure 7: High harmonic emission calculated for ZnO. (a) Shows results where dephasing does not occur; (b) is obtained for a dephasing time of approximately 5 fs. The curve in blue shows two-band (or re-collision) harmonics while the curves in red are for a single band term.

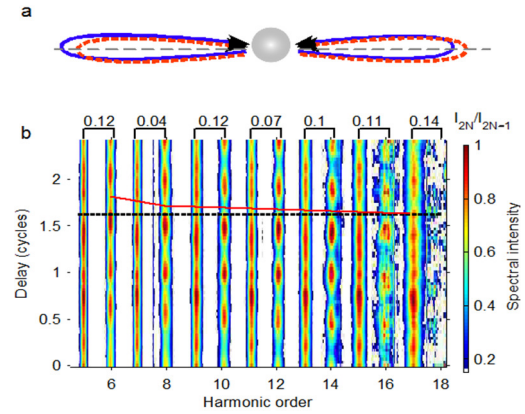


Figure 8: A map of the high harmonic emission from a ZnO sample. The vertical axis shows the delay between the fundamental and second harmonic, the horizontal axis shows the harmonic number and where all data is normalized to the maximum single for that harmonic.

Figure 8 shows that the spectral phase changes with frequency. The measured dependence agrees with that predicted by the two-band (or re-collision) contribution.

There are many consequences of this work. Among them are:

- With a 2-D measurement we can re-construct the band structure of ZnO without otherwise knowing it. (For confirmation, please see below).

- Since we are transferring attosecond technology from gases to solids, measurements can have a time resolution of < 1 femtosecond.

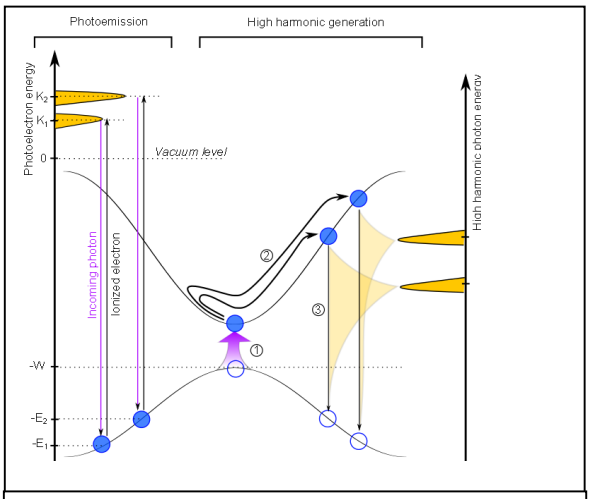


Figure 9: An illustration of a two band model of ZnO. The conventional method to measure band structure is to determine energy and direction of a photoelectron. We propose to use the inverse process. We measure the photon energy of the harmonics to know band gap and the momentum is determined by the two color (insitu) technique.

- High harmonic generation in solids is highly sensitive to weak perturbing fields. This is confirmed by our observation of even harmonics with 2-nd harmonic intensity of only 10^{-5} of the fundamental intensity. This implies that even DC fields in semiconductor circuits imprint their signature onto the emission.
- With short wavelength light produced internal to a material, we can resolve spatial structures up to about $\lambda/2$ of the highest harmonic.

With the underlying physics confirmed, we have tested all optical band reconstruction (as shown in Figure 9 and reported in [4d]). The photon energy of a harmonic determines the band gap at the momentum at which the electron and hole re-collide. If we can determine the momentum at that point, then we have the band differences. Effectively, the momentum is determined by the 2-color map similar to the one shown in Fig. 8

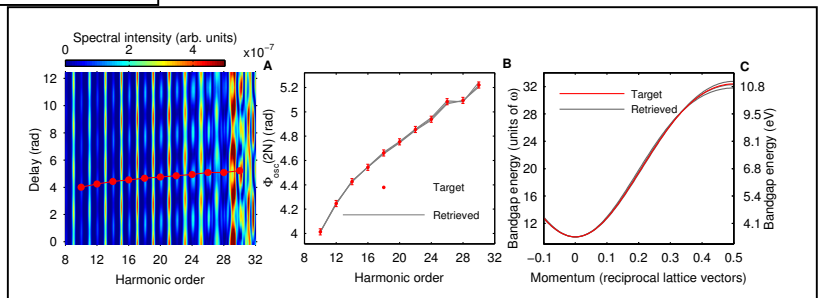


Figure 10: Simulated band gap reconstruction. (A) The simulated experimental data showing the harmonics on the horizontal axis and the relative phase between the fundamental and second harmonic along the vertical. (B) The relative phase where each even harmonic peaks for a retrieved band gap and the initial assumed one (target). (C) The band gap differential as a function of k that we retrieve as well as the initially assumed band structure.

We have implemented this procedure using theoretically generated data, but once obtained, treating the data as if it were experimental. Figure 10 shows the results. The left panel is the simulated two color results. The spectral phase change as a function of frequency is clear from the slope of the red points on the figure, identifying maxima in the even harmonic emission. The middle curve isolates the relative phase between the fundamental and second harmonic as a function of harmonic number. The final panel shows the retrieved curves (black) and the target curve (red). The agreement is almost perfect.

To summarize our conclusions for solids, it is clear that harmonics from solids will have multiple origins. For THz drivers, dephasing will forbid re-collision harmonics. For large band gap dielectrics, localization will likewise forbid re-collision [4c]. However, for a large region between, re-collision seems to play a dominant role.

We believe that this work has established an approach for future experiments in solids, and that gas and solid experiments will begin to integrate. The spectral phase of the different frequencies in the harmonic emission is an important parameter to measure and this parameter can be used to identify the underlying physics. We also believe that this work opens a new route for applying attosecond methods to solids. In the future we should be able to study breakdown in solids with the precision that we now can in gases. This will be important for laser machining.

Total publications: 11 (including 5 in Nature family; 2 in the Physical Review family)

1.

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Dr. Paul Corkum

Program Manager
The AFOSR Program Manager currently assigned to the award

Dr. Enrique Parra

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Abstract

Project goals.
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interferometry, in-situ measurement becomes a comprehensive measurement method.

Aim 2: To exploit few-cycle infrared radiation to create shorter duration attosecond pulses.
Result: We currently reach photon energies > 500 eV but without phase control for the 1.8 μm driver. We obtain CEP control with lower energy driver pulses, but with them we cannot reach 500 eV. Once we combine these, we expect to produce and characterize pulse with durations < 50 as.

Summary of key findings: Every method of generating isolated attosecond pulses requires carrier-envelop-phase control. We generate CEP controlled pulses for 1.8 μm light by using an OPA in which the seed, at the signal wavelength, is generated by continuum generation. In this way the idler is CEP stable. This pulse is then frequency broadened in a hollow core fiber and passed through fused quartz where anomalous dispersion compresses it. We achieve 600 μJ pulses with duration of less than 13 fs. With this source we can produce isolated attosecond pulses. With 600 μJ pulses we can reach the water window using neon gas and a thin jet with high backing pressure.

Aim 3: To compare attosecond gating methods, selecting the most efficient way to produce the highest power or the shortest duration attosecond pulses.
Results: We have adapted the attosecond lighthouse to gases and confirmed isolated attosecond pulse production with an IR driver. We are working on a new method using a solid irradiated at high intensity to create a space-time coupled pulses that then irradiate a gas jet.

Summary of key findings: We have explored two new gating methods. Both are promising. In one we use a fundamental beam that is space-time coupled. We create a beam at the focus that has a wave front that changes from one half cycle to the next. When the fundamental beam is converted to harmonics the higher frequency of the harmonics means that the beam has lower divergence. The attosecond pulses that make up the harmonics, therefore, separate into individual attosecond pulses in the far field. The second gating method also uses a space-time coupled pulse. To create it we use of the response of thin (about 200 μm) crystalline quartz plates to intense light. The resulting space-time coupled, two-color pulse is then incident on a gas target placed some distance away where it creates an isolated attosecond pulse in xenon, krypton and probably all rare gases.

Aim 4: To use the quantum path interference, present in any attosecond pulse, to resolve the strongly-driven attosecond time-scale electron dynamics in selected ions.
Results: This aim has been modified to concentrate on high harmonics generation in solids.

Summary of key findings: We began to understand the deep connection between attosecond pulses made in the gas phase and high harmonics formed in wide bandgap semiconducting material. After discussions with the Program Manager, we modified Aim 4 to study high harmonics generation within solids. By studying a two-band model of wide bandgap semiconductors, we have discovered that, harmonics can have essentially the same origin in solids as in gases. The qualification "essentially" only refers to the natural generalization that electron and hole motion in solids takes the place of electrons and ions in gases and that the electrons and holes move with band-corrected masses in a semiconductor rather than with their real electron and ion mass in gases.

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Archival Publications (published) during reporting period:

1. K.T. Kim, C. Zhang, A.D. Shiner, S.E. Kirkwood, E. Frumker, G. Gariépy, A. Naumov, D.M. Villeneuve and P.B. Corkum, "Manipulation of quantum paths for space-time characterization of attosecond pulses", Nature Physics 9, 159-163 (2013).
2. K.T. Kim, C. Zhang, A.D. Shiner, B.E. Schmidt, F. Legare, D.M. Villeneuve and P. B. Corkum, "Petahertz optical oscilloscope", Nature Photonics, 7, 958-962 (2013).
3. A.D. Shiner, C. Trallero-Herrero, N. Kajumba, B.E. Schmidt, J.B. Bertrand, Kyung Taec Kim, H.-C. Bandulet, D. Comtois, J.-C Kieffer, D.M. Rayner, P.B Corkum, F. Légaré, D.M. Villeneuve, "High harmonic cut-off energy scaling and laser intensity measurement with a 1.8 μm laser source" Journal of Modern Optics, DOI:10.1080/09500340.2013.765067 (2013).
4. J.B Bertrand, H.J. Wörner, P. Salières, D.M. Villeneuve and P.B. Corkum, "Linked attosecond phase interferometry for molecular frame measurements" Nature Physics 9, 174-178 (2013).
5. K.T. Kim, D. M. Villeneuve, P. B. Corkum, "Manipulating quantum paths for novel attosecond measurement methods" Nature Photonics 8, 187-194 (2014).
6. Yijian Meng, Chunmei Zhang, Claude Marceau, A. Yu. Naumov, P. B. Corkum, and D. M. Villeneuve, "Octave-spanning hyperspectral coherent diffractive imaging in the extreme ultraviolet range", Optics Express 23, 28960 (2015).
7. C. Zhang, G. Vampa, D.M. Villeneuve and P.B. Corkum, "Attosecond lighthouse driven by sub-two-cycle, 1.8 μm laser pulses" J. Phys. B.: At. Mol. Opt. Phys. 48, 061001 (2015).
8. T.J. Hammond, K.T. Kim, C. Zhang, D.M. Villeneuve, and P.B. Corkum, "Controlling attosecond angular streaking with second harmonic radiation" Optics Letters, 40, 1768, (2015).
9. G. Vampa, T.J. Hammond, N. Thiré, B.E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec and P.B. Corkum, "Linking high harmonics from gases and solids" Nature 522, 462 (2015)
10. C.R. McDonald, G. Vampa, P.B. Corkum, and T. Brabec, "Interband Block oscillation , Phys. Rev. A 92, 033845 (2015).
G. Vampa, T. J. Hammond, N. Thiré, B. E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec, D. D. Klug, and P. B. Corkum, "All-Optical Reconstruction of Crystal Band Structure", Phys. Rev. Lett. 115, 193603 (2015).

Changes in research objectives (if any):

After discussions with the Program Manager, Aim 4 have been modified to concentrate on harmonics generation in solids.

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

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Appendix Documents

2. Thank You

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